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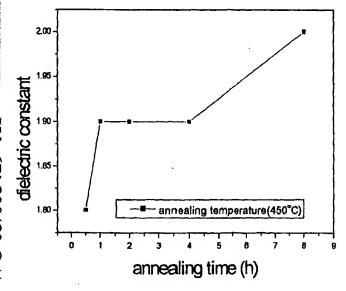
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(54) Title: METHOD FOR PREPARING LOW DIELECTRIC FILMS



(57) Abstract: A low dielectric constant hydrogenated silicon-oxycarbide (SiCO:H) film is prepared by conducting chemical vapor deposition using, together with an O₂-containing gas plasma, an organosilicon or organosilicate compound having at least one vinyl or ethinyl group, or a mixture of a saturated organosilicon or organosilicate compound and an unsaturated hydrocarbon.

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METHOD FOR PREPARING LOW DIELECTRIC FILMS

FIELD OF THE INVENTION

The present invention relates to an improved plasma chemical vapor deposition (CVD) method for preparing a low dielectric constant hydrogenated silicon-oxycarbide (SiCO:H) film.

BACKGROUND OF THE INVENTION

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With ever decreasing size of electronic devices utilized in ULSI (ultra-large-scale integrated) circuits, there has emerged the problem of increased capacitance of intralayers and/or interlayers, causing signal delays.

Therefore, there has been a need to develop a low dielectric constant (k) material having a k-value lower than that of the conventional silicon dioxide (SiO₂) or fluorinated silicon oxide (SiOF).

US Patent No. 6,147,009 discloses a low dielectric constant material produced by reacting the vapor of a precursor containing atoms of Si, C, O and H in a parallel plate plasma enhanced chemical vapor deposition chamber, the 20 precursor being molecule with a ring structure such 1,3,5,7-tetramethylcyclotetrasiloxane(TMCTS, C4H16O4Si4), tetraethylcyclotetrasiloxane(C₈H₂₄O₄Si₄) decamethylcyclopentasiloxane(C₁₀H₃₀O₅Si₅), with or without added oxygen. However, the dielectric constant of the disclosed film is still high, in the range 25 of 3.3 to 4.0. . To further reduce the dielectric constant of the material described in the patent, US Patent No. 6,312,793 proposes a low k material consisting of two or more phases. However, the multi-phase material still has a k-value of more than 3.2.

Accordingly, the present inventors have endeavored to develop a novel 30 material having a dielectric constant lower than those of conventional materials.

SUMMARY OF THE INVENTION

It is, therefore, an object of the present invention to provide a method

for preparing an improved low dielectric constant hydrogenated silicon-oxycarbide (SiCO:H) film using a chemical vapor deposition (CVD) technique.

It is another object of the present invention to provide a SiCOH film 5 having a dielectric constant (k) lower than those of conventional low dielectric materials.

In accordance with one aspect of the present invention, there is provided a method for preparing a low dielectric constant hydrogenated silicon-oxycarbide (SiCO:H) film which comprises conducting chemical vapor deposition using, together with an O₂-containing gas plasma, an organosilicon or organosilicate compound having at least one vinyl or ethinyl group, or a mixture of a saturated organosilicon or organosilicate compound and an unsaturated hydrocarbon.

In accordance with another aspect of the present invention, there is provided a low dielectric constant SiCOH thin film having a dielectric constant (k) of 2.6 or below, prepared by said method.

BRIEF DESCRIPTION OF THE DRAWINGS

The above and other objects and features of the present invention will become apparent from the following description thereof, when taken in conjunction with the accompanying drawings which respectively show:

FIGS. 1a and 1b: schematic diagrams of the plasma reactor used in forming a thin film in accordance with the present invention;

- FIG. 2: variations in carbon contents of the film obtained in Example 1 of the present invention with O₂/VTMS flow ratio, respectively;
 - FIG. 3: variations in dielectric constants of the films obtained in Example 1 and Comparative Example 1 of the present invention with $O_2/VTMS$ and $O_2/4MS$ flow ratio, respectively;
- FIGS. 4 and 5: changes in the dielectric constant of the film obtained in Example 1 as function of annealing temperature and annealing time, respectively;

FIGS. 6 and 7: variations in carbon contents and dielectric constants of the film obtained in Example 2 of the present invention with O₂/(4MS+C₂F₄) 35 flow ratio, respectively;

FIGS. 8 and 9: changes in the dielectric constant of the film obtained in

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Example 2 as function of annealing temperature and annealing time, respectively;

FIG. 10: variations in carbon contents of the film obtained in Example 3 of the present invention with O₂/TVTMCTSO flow ratio, respectively;

FIG. 11: variations in dielectric constants of the films obtained in Example 3 and Comparative Example 2 of the present invention with O₂/TVTMCTSO and O₂/TMCTSO flow ratio, respectively;

FIGS. 12 and 13: changes in the dielectric constant of the film obtained in Example 3 as function of annealing temperature and annealing time, 10 respectively;

FIGS. 14 and 15: variations in carbon contents and dielectric constants of the film obtained in Example 4 of the present invention with O₂/(TMCTSO+C₂H₄) flow ratio, respectively;

FIGS. 16 and 17: changes in the dielectric constant of the film obtained 15 in Example 4 as function of annealing temperature and annealing time, respectively;

FIGS. 18 and 19: variations in carbon contents and dielectric constants of the film obtained in Example 5 of the present invention with O₂/DADMS flow ratio, respectively;

FIGS. 20 and 21: changes in the dielectric constant of the film obtained in Example 5 as function of annealing temperature and annealing time, respectively;

FIGS. 22 and 23: variations in carbon contents and dielectric constants of the film obtained in Example 6 of the present invention with O₂/DVTMDSO 25 flow ratio, respectively;

FIGS. 24 and 25: changes in the dielectric constant of the film obtained in Example 6 as function of annealing temperature and annealing time, respectively:

FIGS. 26 and 27: variations in carbon contents and dielectric constants 30 of the film obtained in Example 7 of the present invention with O₂/VTMOS flow ratio, respectively;

FIGS. 28 and 29: changes in the dielectric constant of the film obtained in Example 7 as function of annealing temperature and annealing time, respectively;

FIGS. 30 and 31: variations in carbon contents and dielectric constants of the film obtained in Example 8 of the present invention with O₂/ETMS flow

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ratio, respectively;

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FIGS. 32 and 33: changes in the dielectric constant of the film obtained in Example 8 as function of annealing temperature and annealing time, respectively;

FIGS. 34 and 35: variations in carbon contents and dielectric constants of the films obtained in Example 9 of the present invention with O₂/(HMDSO+C₂H₄) flow ratio, respectively; and

FIGS. 36 and 37: changes in the dielectric constant of the film obtained in Example 9 as function of annealing temperature and annealing time, 10 respectively.

DETAILED DESCRIPTION OF THE INVENTION

The present invention provides a method for preparing a low dielectric constant hydrogenated silicon-oxycarbide (SiCO:H) film by way of conducting chemical vapor deposition using, together with an O₂-containing gas plasma, an organosilicon or organosilicate compound having at least one vinyl or ethinyl group, or a mixture of a saturated organosilicon or organosilicate compound and an unsaturated hydrocarbon.

In accordance with the present invention, the process for forming a low dielectric constant SiCOH film may be conducted using a plasma CVD apparatus, e.g., a remote plasma CVD or a direct plasma CVD apparatus.

The remote plasma CVD apparatus shown in FIG. 1a comprises a quartz tube(6), an antenna(9), a matching box(5), a high-frequency electric power source(10), a mass flow controller(7) for feeding a silicon precursor, a precursor container(2), and a mass flow controller(8) for feeding a reactive gas. The antenna is wound around the outer periphery of the quartz tube(6), to thereby connect the antenna(9) and the matching box(5), which is connected to the high-frequency electric power source(10). The quartz tube(6) is connected to the mass flow controller(8) for feeding an O₂-containing gas via a tube. In the remote plasma CVD apparatus, the O₂-containing gas and the silicon precursor are fed to the matching box(5) separately, the precursor being led through a diffusion ring(3). Further, in the direct plasma CVD apparatus shown in FIG. 1b, the silicon precursor fed from 35 the precursor container(2) and the O₂-containing gas fed from a reactive gas container(1) are mixed, and then supplied to the matching box(5).

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In accordance with a preferred embodiment of the present invention, there is provided a method for preparing a low dielectric constant SiCOH material, which comprises conducting chemical vapor deposition using an unsaturated organosilicon or organosilicate compound having at least one vinyl 5 or ethinyl group and an O₂-containing gas plasma.

Representative examples of the unsaturated organosilicon organosilicate compound having at least one vinyl or ethinyl group include vinyltrimethylsilane, vinyltrimethoxylsilane, vinyltriethylsilane, 1,3,5-trivinyl-1,3,5-trimethylcyclotrisiloxane, vinyltriethoxylsilane, 10 1,3,5,7-tetravinyl-1,3,5,7-tetramethylcyclotetrasiloxane,

1.3-divinyltetramethyldisiloxane, hexavinyldisiloxane, allyldimethylsilane, allyldimethoxysilane, ethinyltrimethylsilane, ethinyltriethylsilane and a mixture thereof.

The unsaturated organosilicon or organosilicate compound may be 15 generated in situ, using an organosilicon or organosilicate having one or more halogen substituents.

In another preferred embodiment of the present invention, there is provided a method for preparing a low dielectric constant SiCOH material, which comprises conducting chemical vapor deposition using a mixture of a 20 saturated organosilicon or organosilicate compound and an unsaturated hydrocarbon with an O2-containing gas plasma. The mixing ratio of the saturated organosilicon or organosilicate compound and the unsaturated hydrocarbon is preferably in the range from 1:0.1 to 1:10. If the mixing ratio is less than 0.1, the dielectric constant of the film becomes too high, while the 25 physical properties of the film become unsatisfactory if the ratio is above 10.

In the above embodiment, the saturated organosilicon or organosilicate compound, or the unsaturated hydrocarbon may have one or more halogen substituents.

Representative examples of the saturated organosilicon or organosilicate trimethoxysilane, 30 compound trimethylsilane, triethylsilane, include tetramethoxysilane, tetramethylsilane, tetraethylsilane, triethoxysilane, tetraethoxysilane, hexamethylcyclotrisiloxane, tetramethylcyclotetrasiloxane, octamethylcyclotetrasiloxane, tetraethylcyclotetrasiloxane, hexamethyldisiloxane, bistrimethylsilylmethane and a mixture thereof.

Representative examples of the unsaturated hydrocarbon are H₂C=CH₂, 35 F₂C=CF₂, H₂C=CF₂, HFC=CFH, F₂C=C=CF₂, H₂C=C=CF₂, HFC=C=CFH,

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HC CH, FC CH, FC CF, Cl₂C=CCl₂, H₂C=CCl₂, HClC=CClH, Cl₂C=C=CCl₂, H₂C=C=CCl₂, HClC=CClH, ClC CH, ClC CCl, Br₂C=CBr₂, H₂C=CBr₂, HBrC=CBrH, Br₂C=C=CBr₂, H₂C=C=CBr₂, HBrC=C=CH, BrC CBr, I₂C=CI₂, H₂C=CI₂, HIC=CIH, IC CH and IC CI; and preferred is H₂C=CH₂ or F₂C=CF₂.

The O₂-containing gas which may be used in the present invention is selected from the group consisting of O₂, N₂O, O₃, H₂O₂, CO₂, H₂O and a mixture thereof.

The method of the present invention may further comprise the step of annealing the deposited film, which may be carried out at a temperature ranging from 100 to 800°C for a period ranging from 0.5 to 8 hrs, preferably at 450°C for 1 hr, to obtain a thermally stable low dielectric constant SiCOH film.

The annealing step may also include a rapid-thermal processing, which may be conducted at a temperature ranging from 100 to 900°C for about 1 minute and a spike-heating step performed for 10 seconds.

Such low dielectric constant SiCOH material prepared in accordance with the method of the present invention has a dielectric constant (k) of 2.8 or below; and, further, the thermally stable SiCOH film formed after annealing 20 has an exceptionally low dielectric constant (k) in the range of 1.6 to 2.6, the dielectric constant (k) being controllable by adjusting the process variables.

The present invention is further described and illustrated in Examples provided below, which are, however, not intended to limit the scope of the present invention.

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Example 1

A SiCOH film was deposited on a Pt substrate using vinyltrimethylsilane(VTMS, SiC₅H₁₂) and O₂ in the direct plasma apparatus shown in FIG. 1b. The flow ratio of O₂/VTMS was varied in the range of 1 to 13.3 during the film deposition. The pressure and temperature in the reactor were 1mmHg and 30°C, respectively, and the applied plasma power, 60W. The film so deposited was annealed under an Ar atmosphere at a temperature in the range of 300 to 500°C, to obtain a low dielectric constant film.

As shown in FIG. 2, the respective carbon contents of the deposited film and the film annealed at 450°C become lower with the increasing flow

ratio of O₂/VTMS. FIG. 3 exhibits that the annealed film at 450°C has a dielectric constant ranging from 1.8 to 2.4, while the deposited film without the annealing has a dielectric constant ranging from 2.3 to 2.8. FIGS. 4 and 5 show the changes in the dielectric constant of the film obtained in Example 1 with the changes in the annealing temperature and annealing time at the O₂/VTMS flow ratio of 2, respectively.

Comparative Example 1

The procedure of Example 1 was repeated using tetramethylsilane(4MS, 10 SiC₄H₁₂) in place of VTMS, to obtain a deposited film. As shown in FIG. 3, the film thus obtained has a dielectric constant ranging from 3.0 to 3.5, which is higher than that of the deposited film obtained in Example 1.

Example 2

Except that a mixture of tetramethylsilane(4MS, SiC₄H₁₂) and C₂F₄ (1:1) was used instead of VTMS, the procedure of Example 1 was repeated to obtain a deposited film, which was subsequently annealed.

FIG. 6 and FIG. 7 show the carbon contents and dielectric constants of the deposited film and the annealed film, respectively. The deposited film 20 has a dielectric constant of 3.0 or below, and the film annealed at 450°C has a dielectric constant of 2.5 or below. FIG. 8 and FIG. 9 show the effects of the annealing temperature (annealing time=0.5hr) and annealing time (annealing temperature=400°C) at the O₂/(4MS+C₂F₄) flow ratio of 4 on the dielectric constant of the film, respectively. The film annealed at 300 to 25 500°C for 0.5 hr has a dielectric constant of 2.75 or below.

Example 3

The procedure of Example 1 was repeated using tetravinyltetramethylcyclotetrasiloxane(TVTMCTSO, Si₄O₄C₁₂H₂₄) in place of 30 VTMS, to obtain a deposited film, which was subsequently annealed.

FIG. 10 and FIG. 11 show the carbon contents and dielectric constants of the deposited film and the annealed film, respectively. The deposited film has a dielectric constant of 2.4 or below, and the film annealed at 450°C, a dielectric constant of 2.2 or below. FIG. 12 and FIG. 13 show the effects of the annealing temperature (annealing time=0.5hr) and annealing time (annealing temperature=450°C) at the O₂/TVTMCTSO flow ratio of 4 on the

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dielectric constant of the film, respectively. The film annealed at 300 to 500°C for 0.5 hr has a dielectric constant of 2.1 or below.

Comparative Example 2

The procedure of Example 1 was repeated using tetramethylcyclotetrasiloxane (TMCTSO, Si₄O₄C₄H₁₆) in place of VTMS, to obtain a deposited film. As shown in FIG. 11, the film thus obtained had a dielectric constant ranging from 2.5 to 3.3, which is higher than that of the deposited film obtained in Example 3.

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Example 4

Except that a mixture of tetramethylcyclotetrasiloxane (TMCTSO, Si₄O₄C₄H₁₆) and C₂H₄ (1:1) was used instead of VTMS, the procedure of Example 1 was repeated to obtain a deposited film, which was subsequently annealed.

FIG. 14 and FIG. 15 depict the carbon contents and dielectric constants of the deposited film and the annealed film, respectively. The deposited film has a dielectric constant of 2.3 or below, and the film annealed at 450°C has a dielectric constant of 2.2 or below. FIG. 16 and FIG. 17 show the effects of the annealing temperature (annealing time=0.5hr) and annealing time (annealing temperature=400°C) at the (TMCTSO+C₂H₄) flow ratio of 2 on the dielectric constant of the film, respectively. The film annealed at 300 to 500°C for 0.5 hr has a dielectric constant of 2.05 or below.

25 Example 5

The procedure of Example 1 was repeated using diallyldimethylsilane (DADMS, SiC₈H₁₆) in place of VTMS, to obtain a deposited film, which was subsequently annealed.

FIG. 18 and FIG. 19 show the carbon contents and dielectric constants of the deposited film and the annealed film, respectively. The deposited film has a dielectric constant of 2.8 or below, and the film annealed at 450°C, a dielectric constant of 2.4 or below. FIG. 20 and FIG. 21 show the effects of the annealing temperature (annealing time=0.5hr) and annealing time (annealing temperature=450°C) at the O₂/DADMS flow ratio of 4 on the dielectric constant of the film, respectively. The film annealed at 300 to 500°C for 0.5 hr has a dielectric constant of 2.35 or below.

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Example 6

Except that 1,3-divinyltetramethyldisiloxane(DVTMDSO, Si₂OC₈H₁₈) was used in place of VTMS, the procedure of Example 1 was repeated to obtain a deposited film, which was subsequently annealed.

FIG. 22 and FIG. 23 show the carbon contents and dielectric constants of the deposited film and the annealed film, respectively. The deposited film has a dielectric constant of 2.9 or below, and the film annealed at 450°C has a dielectric constant of 2.4 or below. FIG. 24 and FIG. 25 show the effects of the annealing temperature (annealing time=0.5hr) and annealing time (annealing temperature=500°C) at the O₂/DVTMDSO flow ratio of 2 on the dielectric constant of the film, respectively. The film annealed at 300 to 500°C for 0.5 hr has a dielectric constant of 2.15 or below.

15 Example 7

The procedure of Example 1 was repeated using vinyltrimethoxysilane (VTMOS, SiO₃C₅H₁₂) in place of VTMS, to obtain a deposited film, which was subsequently annealed.

FIG. 26 and FIG. 27 show the carbon contents and dielectric constants of the deposited film and the annealed film, respectively. The deposited film has a dielectric constant in the range of 2.2 to 2.75, and the film annealed at 450°C has a dielectric constant in the range of 1.9 to 2.55. FIG. 28 and FIG. 29 show the effects of the annealing temperature (annealing time=0.5hr) and annealing time (annealing temperature=450°C) at the 25 O₂/VTMOS flow ratio of 2 on the dielectric constant of the film, respectively. The film annealed at 300 to 500°C for 0.5 hr has a dielectric constant of 2.4 or below.

Example 8

Except that ethinyltrimethylsilane (ETMS, SiC₅H₁₀) was used in place of VTMS, the procedure of Example 1 was repeated to obtain a deposited film, which was subsequently annealed.

FIG. 30 and FIG. 31 show the carbon contents and dielectric constants of the deposited film and the annealed film, respectively. The deposited 35 film has a dielectric constant of 2.65 or below, and the film annealed at 450°C, a dielectric constant of 2.35 or below. FIG. 32 and FIG. 33 show

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the effects of the annealing temperature (annealing time=0.5hr) and annealing time (annealing temperature=350°C) at the O₂/ETMS flow ratio of 4 on the dielectric constant of the film, respectively. The film annealed at 300 to 500°C for 0.5 hr has a dielectric constant of 2.35 or below.

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Example 9

The procedure of Example 1 was repeated using a mixture of hexamethyldisiloxane(HMDSO, Si₂OC₆H₁₈) and C₂H₄ (1:2) in place of VTMS, to obtain a deposited film, which was subsequently annealed.

FIG. 34 and FIG. 35 show the carbon contents and dielectric constants of the deposited film and the annealed film, respectively. The deposited film has a dielectric constant of 3.0 or below, and the film annealed at 450°C, a dielectric constant of 2.05 or below. FIG. 36 and FIG. 37 show the effects of the annealing temperature (annealing time=0.5hr) and annealing time (annealing temperature=450°C) at the O₂/(HMDSO+C₂H₄) flow ratio of 4 on the dielectric constant of the film, respectively. The film annealed at 300 to 500°C for 0.5 hr has a dielectric constant ranging from 1.7 to 1.9.

As can be seen from the above results, the low dielectric constant SiCOH film prepared by conducting CVD using an O₂-containing gas plasma and an unsaturated organosilicon or organosilicate compound, or a mixture of a saturated organosilicon or organosilicate compound and an unsaturated hydrocarbon in accordance with the present invention has a dielectric constant of 2.6 or below, which is exceptionally lower than conventional low k-materials.

While the subject invention have been described and illustrated with respect to the preferred embodiments only, various changes and modifications may be made therein without departing from the essential concept of the present invention which should be limited only by the scope of the appended claims.

vinyltrimethylsilane,

What is claimed is:

hexamethyldisiloxane,

- 1. A method for preparing a low dielectric constant hydrogenated silicon-oxycarbide (SiCO:H) film which comprises conducting chemical vapor 5 deposition using, together with an O₂-containing gas plasma, an organosilicon or organosilicate compound having at least one vinyl or ethinyl group, or a mixture of a saturated organosilicon or organosilicate compound and an unsaturated hydrocarbon.
- 10 2. The method of claim 1, wherein the mixture of a saturated organosilicon or organosilicate compound and an unsaturated hydrocarbon has a mixing ratio ranging from 1:0.1 to 1:10.
- 3. The method of claim 1, wherein the organosilicon or organosilicate compound having at least one vinyl or ethinyl group is selected from the group consisting of vinyltrimethylsilane, vinyltriethylsilane, vinyltriethoxylsilane, vinyltriethoxylsilane, 1,3,5-trivinyl-1,3,5-trimethylcyclotrisiloxane, 1,3,5,7-tetravinyl-1,3,5,7-tetramethylcyclotetrasiloxane, 1,3,divinyltetramethyldisiloxane, hevavinyldisiloxane, allyldimethylsilane
- 1,3-divinyltetramethyldisiloxane, hexavinyldisiloxane, allyldimethylsilane, 20 allyldimethoxysilane, ethinyltrimethylsilane, ethinyltriethylsilane and a mixture thereof.
- 4. The method of claim 3, wherein the organosilicon or organosilicate compound having at least one vinyl or ethinyl group is selected from the group consisting of vinyltrimethylsilane, 1,3,5,7-tetravinyl-1,3,5,7-tetramethylcyclotetrasiloxane, allyldimethylsilane, 1,3-divinyltetramethyldisiloxane, vinyltrimethoxylsilane and ethinyltrimethylsilane.
- 30 5. The method of claim 1, wherein the saturated organosilicon or organosilicate compound is selected from the group consisting of trimethylsilane, triethylsilane, trimethoxysilane, triethoxysilane, tetramethylsilane, tetramethoxysilane, tetramethoxysilane, tetramethylcyclotetrasiloxane, tetramethylcyclotetrasiloxane, octamethylcyclotetrasiloxane,

bistrimethylsilylmethane,

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vinyltriethylsilane, vinyltrimethoxylsilane, vinyltriethoxylsilane, 1,3,5-trivinyl-1,3,5-trimethylcyclotrisiloxane,

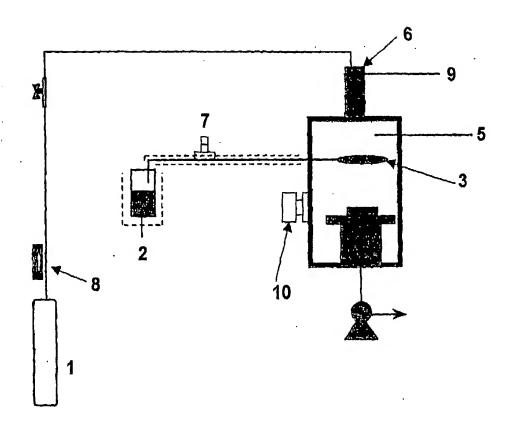
- 1,3,5,7-tetravinyl-1,3,5,7-tetramethylcyclotetrasiloxane,
- 1,3-divinyltetramethyldisiloxane, hexavinyldisiloxane, allyldimethylsilane, 5 allyldimethoxysilane, ethinyltrimethylsilane, ethinyltriethylsilane and a mixture thereof.
- 6. The method of claim 5, wherein the saturated organosilicon or organosilicate compound is selected from the group consisting of 10 tetramethylsilane, hexamethyldisiloxane and tetramethylcyclotetrasiloxane.
- 7. The method of claim 1, wherein the unsaturated hydrocarbon is selected from the group consisting of H₂C=CH₂, F₂C=CF₂, H₂C=CF₂, HFC=CFH, F₂C=C=CF₂, H₂C=C=CF₂, HFC=C=CFH, HC CH, FC CH, FC CF, 15 Cl₂C=CCl₂, H₂C=CCl₂, HCIC=CCIH, Cl₂C=C=CCl₂, H₂C=C=CCl₂, HCIC=CCH, Cl₂C=CBr₂, H₂C=CBr₃, HBrC=CBrH, Br₂C=C=CBr₂, H₂C=C=CBr₃, HBrC=CBrH, Br₂C=C=CBr₂, H₂C=C=CI₂, HIC=C=CIH, I₂C=CI₂, H₂C=CI₂, HIC=C=CIH, I₂C=CI₂, H₂C=C=CI₂, HIC=C=CIH, I₃C=CII₄, IC CH and IC CI.

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- 8. The method of claim 7, wherein the unsaturated hydrocarbon is $H_2C=CH_2$ or $F_2C=CF_2$.
- 9. The method of claim 1, wherein the O₂-containing gas is selected from 25 the group consisting of O₂, N₂O, O₃, H₂O₂, CO₂, H₂O and a mixture thereof.
 - 10. The method of claim 1 further comprising the step of annealing the deposited film at a temperature ranging from 100 to 500°C for 0.5 to 8 hrs.
- 30 11. A low dielectric constant hydrogenated silicon-oxycarbide (SiCO:H) film prepared by the method of claim 1.

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FIG. 1a



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FIG. 1b

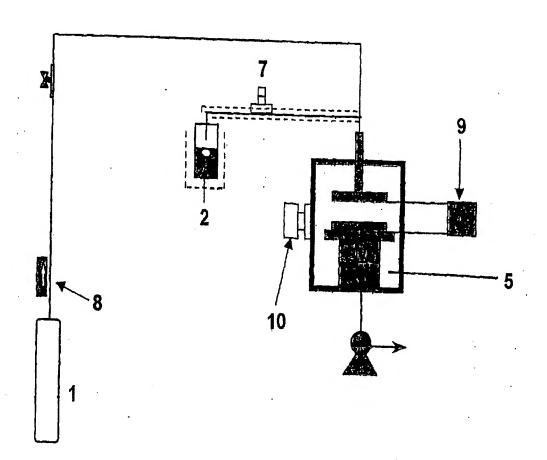


FIG. 2

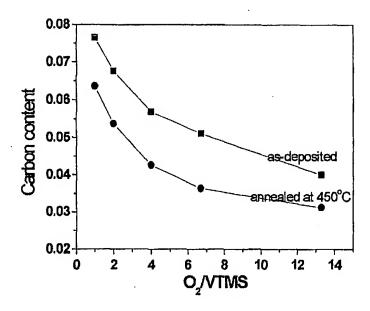
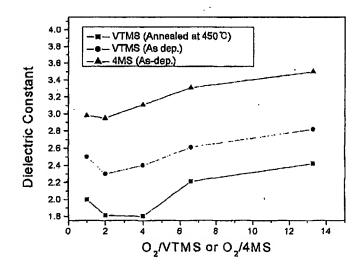


FIG. 3



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FIG. 4

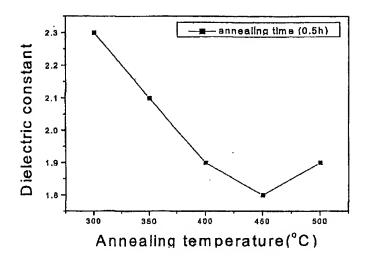
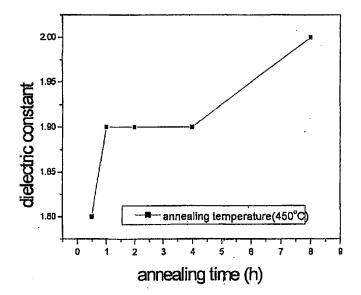


FIG. 5



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FIG. 6

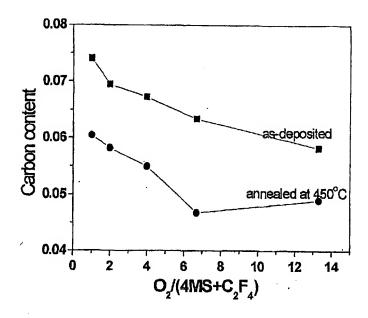


FIG. 7

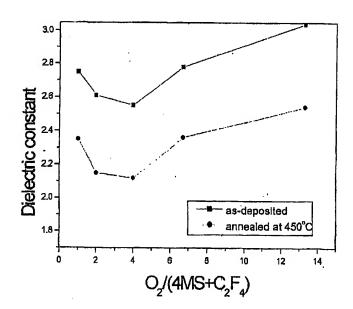


FIG. 8

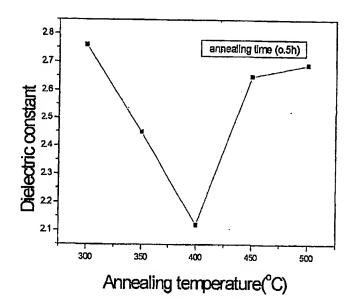
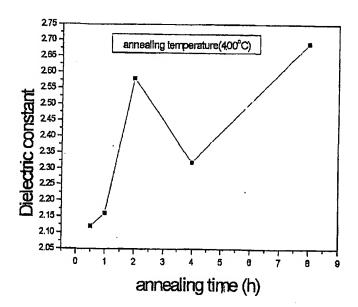


FIG. 9



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FIG. 10

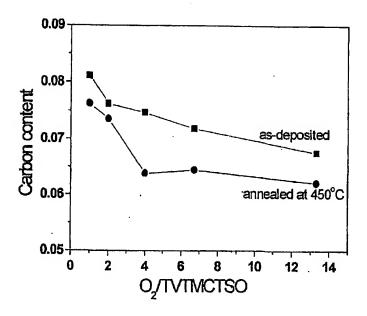


FIG. 11

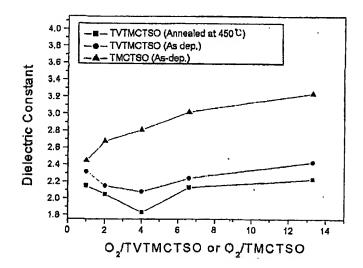


FIG. 12

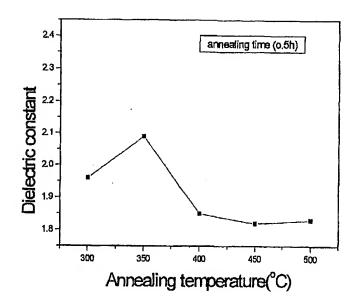
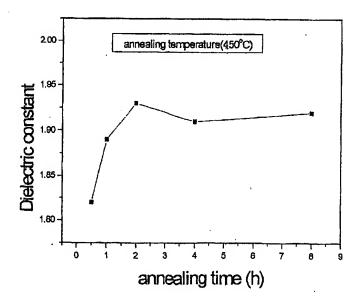


FIG. 13



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FIG. 14

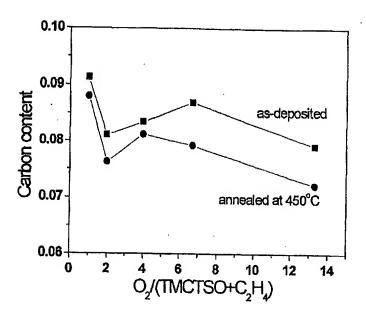


FIG. 15

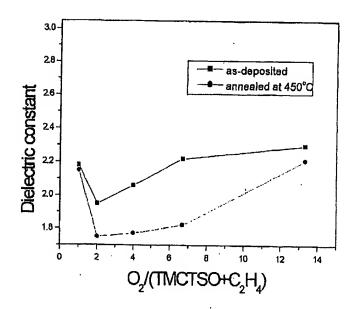


FIG. 16

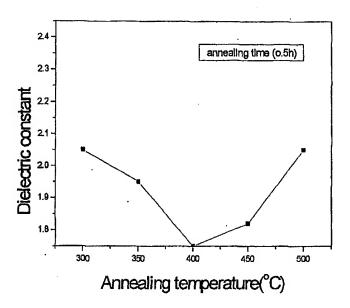
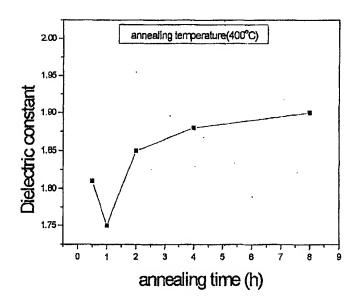


FIG. 17



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FIG. 18

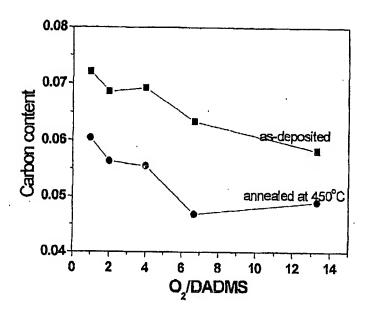
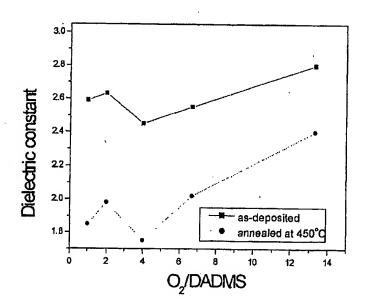


FIG. 19



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FIG. 20

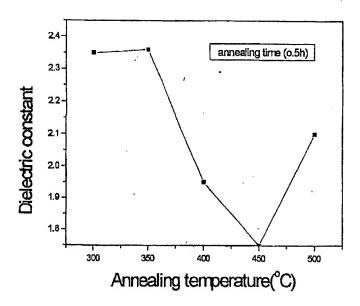
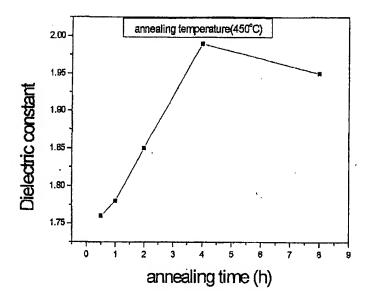
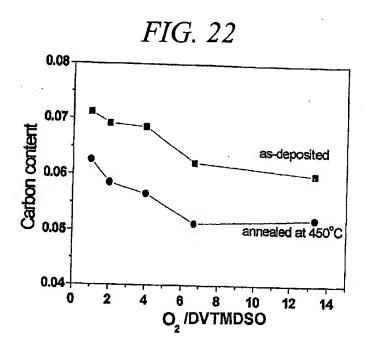
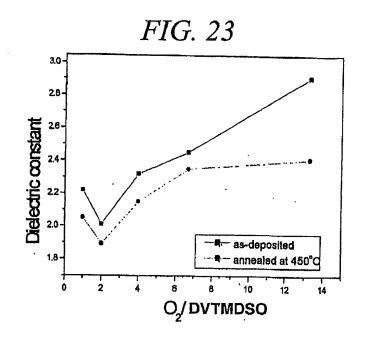


FIG. 21







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FIG. 24

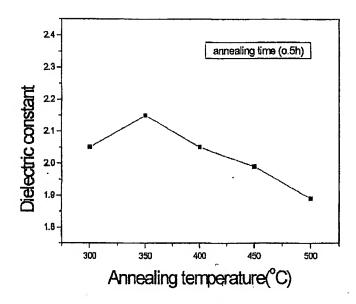
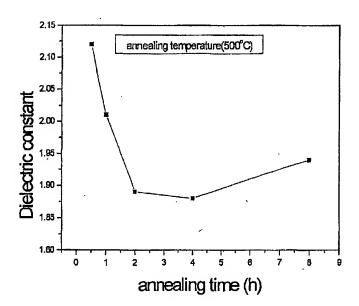


FIG. 25



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FIG. 26

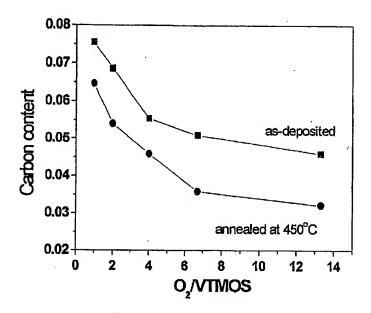
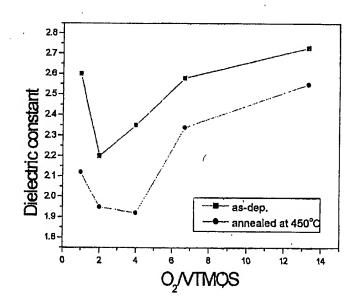


FIG. 27



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FIG. 28

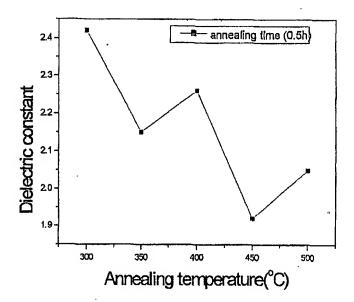
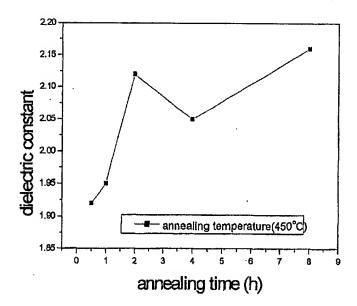
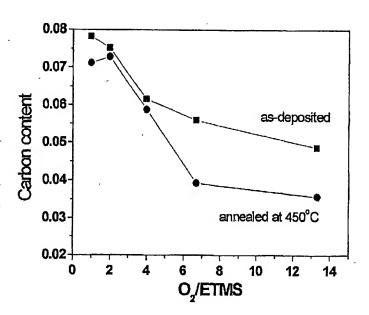


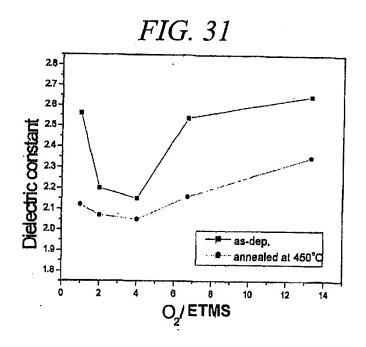
FIG. 29



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FIG. 30





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FIG. 32

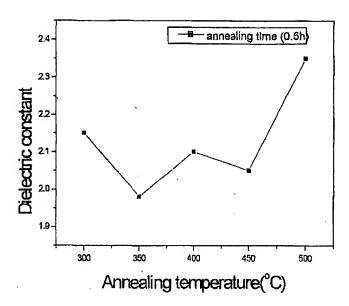
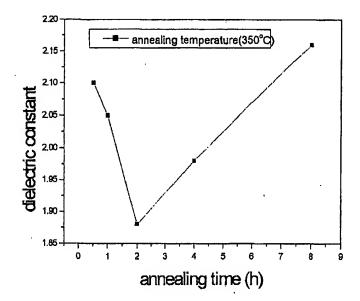


FIG. 33



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FIG. 34

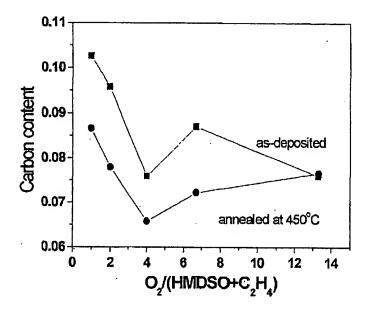
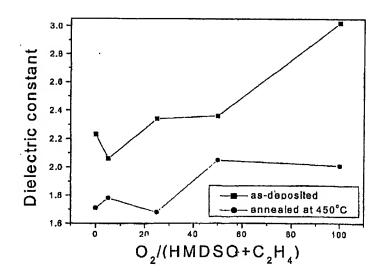


FIG. 35



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FIG. 36

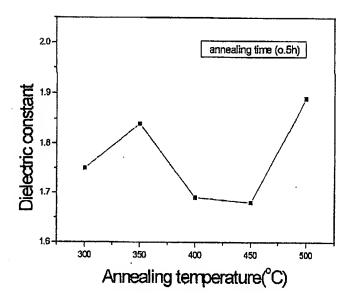
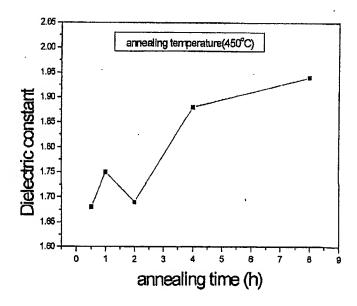


FIG. 37



INTERNATIONAL SEARCH REPORT

International application No.
PCI/KR02/01238

A. CLASSIFICATION OF SUBJECT MATTER			
IPC7 H01L 21/22			
According to International Patent Classification (IPC) or to both national classification and IPC			
B. FIELDS SEARCHED			
Minimum documentation searched (classification system followed by classification symbols)			
PC7 C23C 16/32, C23C 16/30, B32B 3/00			
Documentation searched other than minimum documentation to the extent that such documents are included in the fields searched			
Electronic data base consulted during the intermational search (name of data base and, where practicable, search terms used)			
USPAT, FPD, PAJ			
C. DOCUMENTS CONSIDERED TO BE RELEVANT			
Category*	Citation of document, with indication, where app	propriate, of the relevant passages	Relevant to claim No.
Y	EP, A, 0 960 958 (DOW CORNING CORP.)		1, 3, 4, 5, 6, 11
	01 December 1999 See page 2, line 28 - line 35, page 2, line 48 - page, 3	, line 1	•
Y	WO, A, 99/41423 (APPLIED MATERIALS INC.)		1, 3, 4, 5, 6, 11
10 February 1999 See page 4, line 15 - line 25, page 7, line 15 - page 8,		line 24	
	page 9, line 20 - page 10, line 20		
			·
Y	US, A, 6 147 009 (IBM)	•	ı
	14 November 2000 See the whole document		
			i i
. Y	US A, 6 077 574 (NEC CORP.)		•
	20 June 2000 See the whole document		
	Soo the whole abstraction		;
Further documents are listed in the continuation of Box C. See patent family annex.		See patent family annex.	
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"E" earlier app	pplication or patent but published on or after the international "X" document of particular relevence; the claimed invention cannot be considered novel or cannot be considered to involve an inventive		
	document which may throw doubts on priority claim(s) or which is step when the document is taken alone cited to establish the publication date of citation or other "Y" document of particular relevence; the claimed invention		ned invention connot be
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"O" document referring to an oral disclosure, use, exhibition or other means		combined with one or more other such documents, such combination being obvious to a person skilled in the art	
"P" document published prior to the international filing date but later "&" document member of the same patent family than the priority date claimed		у	
Date of the actual completion of the international search		Date of mailing of the international search report	
29 AUGUST 2002 (29.08.2002)		29 AUGUST 2002 (29.08.2002)	
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